LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-JC-152384

Hydrodynamic Issues in PAMS Mandrel Target Fabrication

B. W. McQuillan, R. Paguio, P. Subramanian, M. Takagi, A. Zebib

August 27, 2003

2003 Third International Conference on Inertial Fusion Sciences and Applications, Monterey, CA
September 7-12, 2003

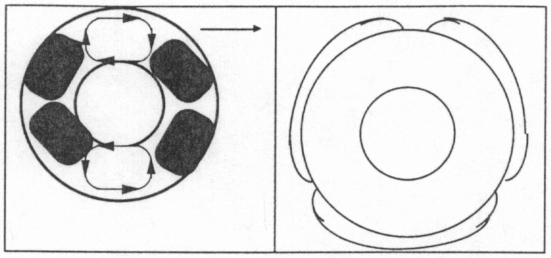

Fig. 2. Image of convection cells and resulting "mode 3" bumpiness.

The convection cells only form if a combination of parameters exceed a critical value Mc

$$M = (d\gamma/dC)(dC/dL)\, L^2/\eta\, D > M_C \quad (1)$$

where $d\gamma/dC$ is the concentration gradient of the outer interfacial surface tension, dC/dL is the radial change of concentration, L is the thickness of he layer, η is the viscosity of the polymer solution, and D is the diffusivity of fluorobenzene in the polymer solution. By adding a bubbler with fluorobenzene, to bubble air with fluorobenzene over the solution of encapsulated shells, we slowed the curing rate and decreased dC/dL, thereby lowering M below Mc. Figure 3 shows how this addition of a bubbler resulted in a much reduced bumpiness across modes 2–20, reduced well below the required NIF standard.

Calculations are underway, to model the Marangoni mode structure throughout the curing process, from first principles. Initial linear theory calculations show that the concentration gradient across the shell wall is very small

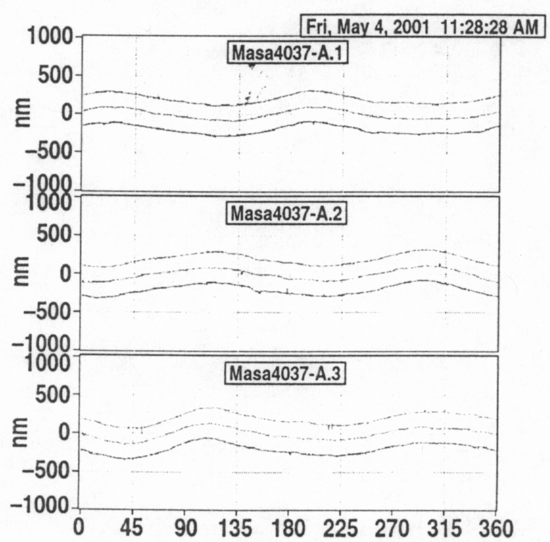

Fig. 3. Shows the shell scans. The resulting power spectrum is shown as the lower red curve in Fig. 1(b).

(ΔC of order 0.1% or less of the average concentration), which confirms the lack of formation of an outer skin, so the outer surface can move toward the inner radius during curing. In an initial calculation with a linear stability model, where "a single 2 mm shell" loses fluorobenzene to an infinite bath near saturation (Cinf = 0.014, where saturation is 0.0150), with a Biot number of 1.0, the stable modes slowly evolve in time from 1+1 to 5 (Fig. 4).

III. A PUZZLE WITH CENTERING

Norimatsu has proposed a model for the centering of the inner water drop within the oil drop.[2] The normal mode oscillation of the compound drop pumps oil (shell wall solution) toward the thin regions, thus pushing the water drop toward the center. We cannot say this model is incorrect, nor can we propose an alternative possibility, but we have observed some data which cannot be explained by his model.

Typically, we add NH_4Cl to the exterior W2 solution (which has PVA), in order to suppress vacuoles and provide density matching in 1 mm shells.[3,4] With 2 mm shells, the suspending W2 has PAA (polyacrylic acid), rather than PVA, and NH_4Cl is incompatible with PAA.[5] We had sought a salt compatible with PAA, and disodium phosphate is compatible. In seeking to improve 1 mm shells, we have attempted adding some PAA (and phosphate) to the standard PVA solution. HOWEVER, the nonconcentricity of the resulting mandrels has consistently been worse, and out of specifications. Figure 5 shows one example, where six beakers of 1 mm shells were made (three at the reference 48°C, and 3 at 60°C). We find these results befuddling: why should the addition of salt on the outside of the shell, effect the motion of the interior water ball? We can surmise a change in interfacial tension, but we have not measured the interfacial tension. A decrease in outside interfacial tension may make the oscillation magnitude larger, thus hastening the centering. So possible

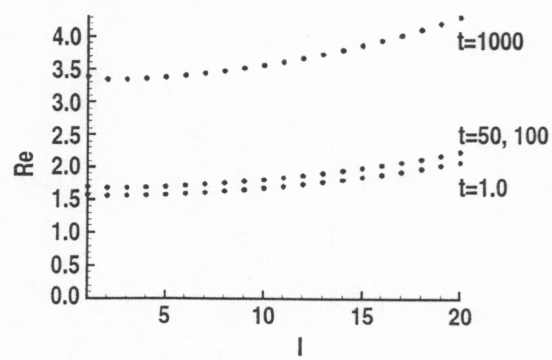

Fig. 4. Marangoni number of each l mode, as a function of time, for a 2 mm shell curing. Most stable modes are 1,2,2,5 at time = 1,50,100,1000 s.

TUP01.38

HYDRODYNAMIC ISSUES IN PAMS MANDREL TARGET FABRICATION

B.W. McQuillan,[1] R. Paguio,[1] P. Subramanian,[2] M. Takagi,[3] and A. Zebib[2]

[1]General Atomics, P.O. Box 85608, San Diego, California 92186-5608 USA
email: barry.mcquillan@gat.com
[2]Mechanical & Aerospace Engineering, Rutgers University, Piscataway, New Jersey 08854-8058 USA
[3]Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, California 94550 USA

Imperfections in PAMS mandrels critically govern the quality of final ICF targets. Imperfections in the mandrels can have a wide range of origins. Here, we present observations of 3 types of imperfections, and data to support the proposal that hydrodynamic factors during the curing of the mandrel are potential causes of these imperfections.

I. INTRODUCTION

The surface finish of a full thickness ICF target depends on the initial symmetry of the PAMS mandrel upon which the ablator layer was coated. Long wavelength surface modulations are reproduced the final coated shell while short wavelength bump defects generally grow in width during the coating process. The surface finish and symmetry requirements for target quality NIF capsules are exceptionally demanding, thus we are focusing significant resources on perfecting the microencapsulation process that is used to produce the initial PAMS mandrel.

The origins of many of the flaws of PAMS mandrels are puzzling to explain and thereby control. We now understand that the simple picture of solvent leaving the shells by diffusion is a flawed model. Complex fluid dynamics plays a critical role, and can be the origin of defects as well as the source of the high level of symmetry that we observe.

II. MODE 10 BUMPINESS

For several years, the surface of 2 mm PAMS mandrels were covered with bumps with an l mode of about 9–10 (Fig. 1). The origin of these bumps was puzzling. Ultimately, their originhas been proposed as originating in Marangoni convection.[1] As fluorobenzene leaves the shell wall during curing, convecton cells are formed, which pump polymer to the outer surface and form bumps (Fig. 2).

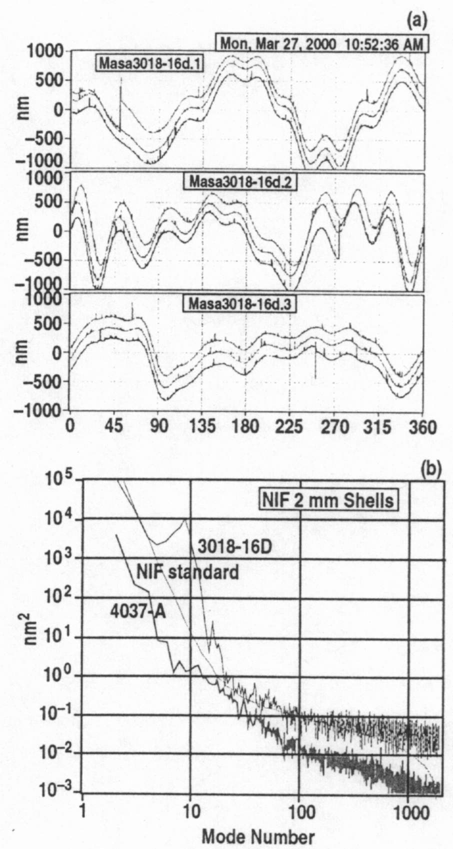

Fig. 1. The scans in 1(a) are shown in blue in 1(b).

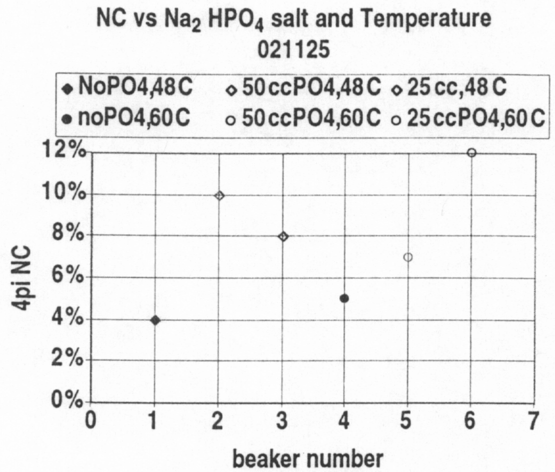

Fig. 5. Beakers 1–3 were at 48°C, 4-6 were at 60°C. Beakers 1 and 4 had no phosphate.

the interfacial tension is increased upon salt addition, decreasing the oscillation magnitude. Alternatively, the salt may complex an impurity in the oil layer, removing the impurity from the inner interface and changing the inner interfacial tension.

IV. ISOLATED DIP

In some (but not all) spheremapper scans of mandrels, there is a localized singular "dip" (Fig. 6). This dip is typically about 0.5–1.0 μm deep, and 20–40° wide. This dip is actually not a dip in the mandrel surface, but a small change in the radius of curvature over a particular region. This local dip results in a large broad bump in the middle modes of the power spectrum above the NIF specification.

The origin of this bump is unknown. In some mandrels we studied closely, there is one significant vacuole beneath the surface of the dip. Whether this vacuole is the cause of the change in radius of curvature, perhaps by changing the flow pattern near the surface, is unknown. Alternatively, a local change in the interfacial tension would also change the radius of curvature. The cause of such a change in interfacial tension is unknown.

In summary, PAMS mandrels made by micro-encapsulation show several imperfections, whose origins would seem to be in fluid flow effects in the mandrels as they cure. Understanding and modeling these hydro-dynamics effects are necessary for making intelligent process changes, for future ICF and IFE targets.

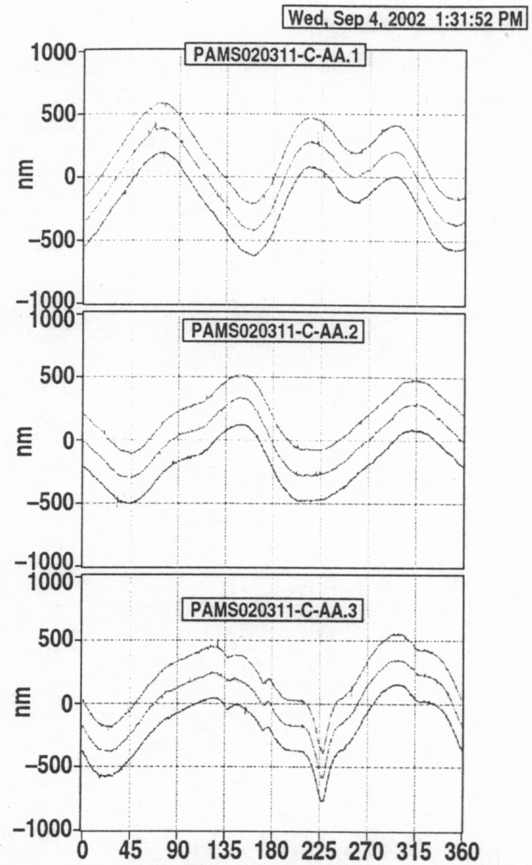

Fig. 6. The isolated dip is seen on the bottom three scans, at about 225 degrees.

ACKNOWLEDGEMENTS

This work was performed under the auspices of the U.S. Department of Energy by General Atomics under contract DE-AC03-95SF20732.and by the University of California Lawrence Livermore National Laboratory under contract W-7405-Eng-48. Work at Rutgers has been supported by NSF grant CTS-0211612.

REFERENCES

1. B.W. MCQUILLAN and M. TAKAGI, "Removal of Mode 10 Surface Ripples in ICF PAMS Shells," *Fusion Science and Technology*, **41**. 209, (2002).

2. T. NORIMATSU, Y. IZAWA, K, MIMA, and P.M. GRESHO, "Modeling of the Centering Force in a Compound Emulsion to Make Uniform Plastic Shells for Laser Fusion Targets," *Fusion Technology* **35**, 147, (1999).

3. B.W. MCQUILLAN and A. GREENWOOD, "Microencapsulation Process Factors Which Influence the Sphericity of 1 mm o.d. Poly(α-Methylstyrene) Shells for ICF," *Fusion Technology*, **35**, 194, (1999).

4. B.W. MCQUILLAN, F.H. ELSNER, R.B. STEPHENS and L.C. BROWN, "The Use of $CaCl_2$ and Other Salts to Improve Surface Finish and Eliminate Vacuoles in ICF Microencapsulated Shells," *Fusion Technology*, **35**, 198, (1999).

5. M. TAKAGI, R. COOOK, R. STEPHENS, J. GIBSON, and S. PAGUIO, "Decreasing Out-Of-Round in Poly(α-Methylstyrene) Mandrels by Increasing Interfacial Tension," *Fusion Technology*, **38**, 46, (2000).